NEURAL NETWORKS IN ANALYSING ^{137}Cs BEHAVIOUR IN THE AIR IN THE BELGRADE AREA

by

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The application of the principal component analysis and artificial neural network method in forecasting ^{137}Cs behaviour in the air as the function of meteorological parameters is presented. The model was optimized and tested using ^{137}Cs specific activities obtained by standard gamma-ray spectrometric analysis of air samples collected in Belgrade (Serbia) during 2009-2011 and meteorological data for the same period. Low correlation (r = 0.20) between experimental values of ^{137}Cs specific activities and those predicted by artificial neural network was obtained. This suggests that artificial neural network in the case of prediction of ^{137}Cs specific activity, using temperature, insolation, and global Sun warming does not perform well, which can be explained by the relative independence of ^{137}Cs specific activity of particular meteorological parameters and not by the ineffectiveness of artificial neural network in relating these parameters in general.

Key words: neural network, gamma-ray spectrometry, air, ^{137}*Cs*

INTRODUCTION

Revealing patterns in vast amount of data obtained from routine monitoring of activities of anthropogenic and natural-occurring radioisotopes requires application of advanced statistical analysis and machine-learning methods. Artificial neural networks (ANN) gained in popularity because of their robustness which outperforms that of conventional statistical techniques. They belong to non-linear methods where no assumption of relations between input variables is needed. A number of ANN algorithms have been developed [1-6] for various purposes. In the area of radioactivity, ANN have been applied in automatic identification of radioactive isotopes in gamma spectroscopy [7], in determination of parameters in monitoring uranium enrichment [8], in portable systems for rapid identification of radionuclides [9-11], in experimental nuclear structure physics (for the gamma ray tracking technique) [12], to optimize parameters of the gamma-spectrometric analysis, and in monitoring radioactive contamination of the environment [13-18].

The ^{137}Cs ($t_{1/2}$ = 30.14 year) is a man-made radionuclide which represents a hazard to the environment. Because of its long half-life ^{137}Cs is regarded as an indicator of anthropogenic pollution caused by nuclear tests and nuclear power plant accidents. Once emitted into the atmosphere it participates in the air-mass circulation processes and can be used for description of pollutant scavenging by precipitation, atmospheric deposition patterns of airborne contaminants, *etc*. [19].

In this study we measured specific activities of ^{137}Cs in near-ground air and correlated them with local meteorological conditions since those have the greatest influence on the stability of the ground layer of atmosphere. Similar to our previous study on ^{7}Be [20], we tested the ability of ANN to predict specific activities of ^{137}Cs in near-ground air by using meteorological parameters as input.

EXPERIMENTAL ANALYSIS

Sampling

Air sampling was conducted in the period from March 2009 to December 2011 at the Kumodraž location, Belgrade, Serbia. It was performed with two digital

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samplers DH 604EV.2 (F&J Speciality Products, Inc. Ocala, Fla., USA) which provide the air flow in the range of 15-120 m^3/h, with an initial air flow rate of 50 ± 5 m^3/h. The air samples were collected weekly with a sampling time of six days at the height of 124 cm from the ground, with a total of 101 samples. Digital samplers were used for measurement of temperature, pressure and relative humidity, while values for wind speed, insolation (sunny hours per day), global Sun warming and precipitation were taken from annual reports of the Republic Hydrometeorological Service of Serbia [21].

Cellulose filter paper FJ213340 1.770 mm thick with an efficiency of 65% on the dioctyl phthalate (DOP) test was used. The DOP test was used to determine the efficiency of the filter with an area of 100 cm^2 for air or gas filtration with the flow rate of 32 L/min containing DOP particles at a concentration of 100 mg/L [22].

Gamma-ray spectrometric analysis

The measurements were performed using a gamma-ray spectrometric system AMETEK-AMT (ORTEC, USA) with a coaxial high-purity germanium (HPGe) detector with relative efficiency of 59.2% measured on the line 1.33 MeV ^{60}Co. Resolution of the device was 1.78 keV on the line 1.33 MeV ^{60}Co. The detector was housed in home-made lead casing. Lead protection was 11 mm thick and covered with a copper sheet 5 mm thick.

Gamma-ray spectrometric analyses were conducted on the device calibrated to the filter geometry. The solution used for calibration was obtained by dilution of the reference IAEA material (^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co). The standard was made by dripping the radioactive solution on the circular filter paper in the hexagonal network. Fifty-five points were applied, each with a volume of 10 μL. ^{137}Cs specific activity of air samples was evaluated from its line at 661 keV, with a relative standard deviation that ranged from 8.8% to 13.7%, for 250 000 s by using the Gamma Vision 32 software package [23].

Data analysis

Principal component analysis (PCA). The extraction of major factors of variations within activity data was performed using PCA (SPSS v 13 software package, SPSS Inc., Chicago, USA) [24] on the set of gamma spectra acquired during 2010. PCA is a statistical method which allows identification of major factors within a multidimensional data set in order to reduce the number of variables of a given data set. Commonly, it is used when there are a great number of correlated variables within one data set. The factors were tested on the correlation with meteorological parameters; the parameters corresponding to the highest correlation coefficients were selected and used as input for the ANN analysis.

Artificial neural network. The most common type of ANN used in the analysis of environmental samples is the multilayer perceptron. Here, the three-layer feed-forward network with the back-propagation algorithm was used based on our previous experience in the neural network application in gamma-ray spectrometry [14-16].

The training process of this network was performed in two phases. In the first phase, the data contained in the input layer were sent to the hidden layer through the input nodes. Nodes in the hidden layer calculate the weight sums of input data. All these sums were then sent to the output layer through the activation functions as single end data.

In our case, we used the logistic function as an activation function which is a representative of sigmoid activation functions

$$f_j = \frac{1}{1 + \exp\left(-\sum w_{ji} o_i - b\right)} \tag{1}$$

where w_{ji} is the weight factor whose value connects the lower layer node i to the upper layer node j, o_j – the output value of the node i, while b represents a bias. The bias (limit of neural activation) is used to calculate the total numerical value of all nodes located in the single layer.

In the second phase of network training, the error was calculated between the calculated and experimentally obtained values in the output layer using the so-called general delta rule. Based on this rule, nodes in the output layer are adjusted according to the value of nodes in the input layer based on the equation

$$w_{ji}^{n+1} = w_{ji}^{n} + \eta\delta_j o_j + \alpha w_{ji}^{n} \tag{2}$$

where δ_j is the signal error at node j, o_j – the output value of node j, n – the number of iterations, η – the learning rate, and α – the momentum. The learning rate affects the step size in the space of weight coefficients and controls the speed at which the network learns. Momentum is included in the calculation to add the previous changes of weight coefficients to the current change during the training process. In determining the value of the learning rate and momentum, we used previous experience in the application of neural networks in gamma-spectrometry and determined that both these values are 0.1 [16].

The key part of the calculation in artificial neural networks is forming the sets for training and testing the network. The training set is used to optimize the model characteristics, while the testing set is used to assess the generalization capabilities of a given network. The training set contained logarithmic values of meteorological parameters as input data and those of ^{137}Cs specific activity from 2009 as output data. The same parameters from 2010 were used as input and output data in the testing set.

The optimal number of nodes in the hidden layer was found by calculating the RMSE (root mean square error) for different values of nodes, from 2 to 22. This was performed for different number of epochs: 1000, 5000, 10000, 20000, 30000, 40000, and 50000.

In order to check the network operation, another data set is formed, called the validation set, whose primary purpose is to assess the formed network. It consisted of the logarithmic values of the meteorological parameters from 2011 as input data and those of ^{137}Cs activity in the same period as output data. This last part is called the cross-validation process [25].

To determine the appropriate number of epochs and to verify the values of network parameters, the validation data set was inserted in the network. The validation set is used to determine if the so-called *over-fitting* of the network has occurred during training. When applying the methods of early stopping, the network is inserted with the validation set instead of the testing set, and the validation error is calculated periodically. When the validation error, after noticeable convergence, starts to grow again, the training process is stopped.

The network optimization process is followed by its testing. The testing set contained 38 measurements of the ^{137}Cs activities, obtained during 2010. Validation of network predictive ability is performed by comparing the values of specific activities obtained experimentally and those calculated using the neural network. Analysis of behaviour of the specific ^{137}Cs activities by using neural networks was performed in the software package QwickNet 2.23 [26].

RESULTS

The ^{137}Cs average monthly activities for the period from March 2009 to December 2011 are given in fig. 1.

Table 1 shows statistical characteristics for all the experimental data for the given period. Meteorological parameters were selected using the PCA. Data from 2009 were used for creating the training set, while data from 2010 and 2011 were used for creating the testing and validation sets, respectively.

The results presented in fig. 2 show that the optimal number of nodes in the hidden layer of the ANN

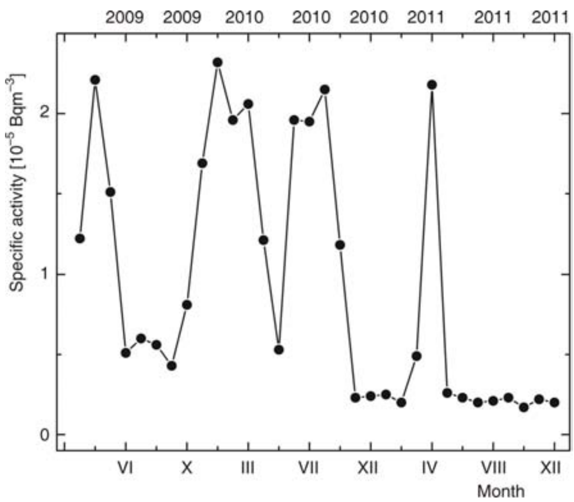

Figure 1. The ^{137}Cs average monthly activity for the period from March 2009 to December 2011 measured at Kumodraž, Belgrade

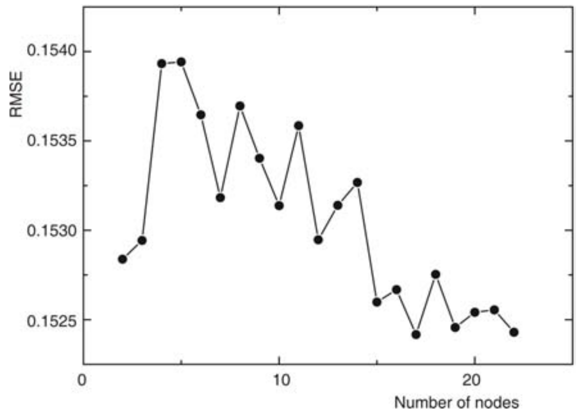

Figure 2. RMSE dependency on the number of nodes in the hidden layer

was 17, because RMSE had the smallest value for this number of nodes.

Figure 3 presents the RMSE for training, testing and validation sets of the model depending on the number of epochs. Results converged and the value of 40000 epochs was considered as optimal.

Schematic representation of the optimized network is shown in fig. 4. The input layer is presented by three nodes consisting of meteorological parameters and the output layer is presented by one node-^{137}Cs activities. The network structure also consists of biases

Table 1. Descriptive statistics of experimental data for the period from march 2009 to December 2011

	2009		2010		2011	
Experimental data	Mean	Min/Max	Mean	Min/Max	Mean	Min/Max
A_C [10^{-5} Bqm^{-3}]	1.0 0.7	0.4/2.5	1.4 0.9	0.2/2.4	0.4 0.6	0.09/3.7
Temperature [°C]	25 6	8.3/34.5	18 10	–0.6/34.7	22.0 9.5	0.8/36.1
Insolation [h]	46 19	6.1/74.9	28 18	0.0/74.1	42 23	0.0/88.3
Global Sun warming [Wcm^{-2}]	1625 605	301/2615	915 621	213/2320	1285 688	114/2475

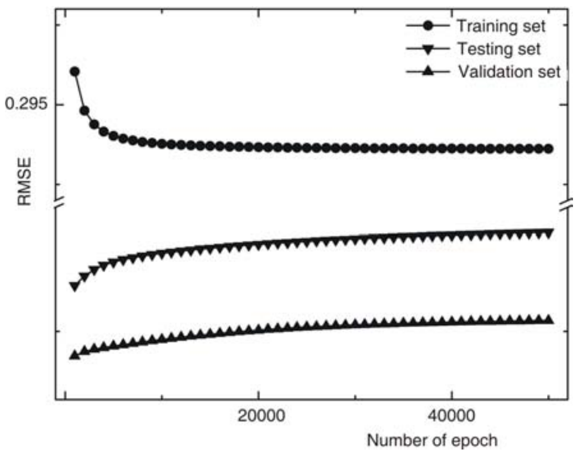

Figure 3. RMSE for training, testing, and validation sets of model dependency on the number of epochs

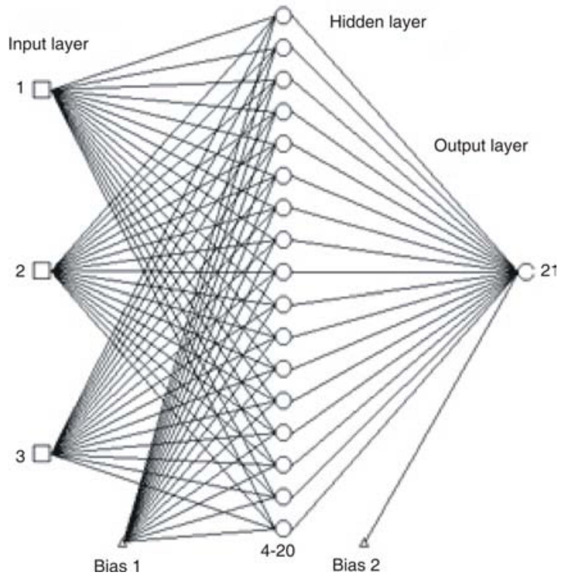

Figure 4. Schematic representation of the optimized three-layer feed-forward neural network used in this study: input layer – nodes 1-3; hidden layer – nodes 4-20, and output layer – node 21

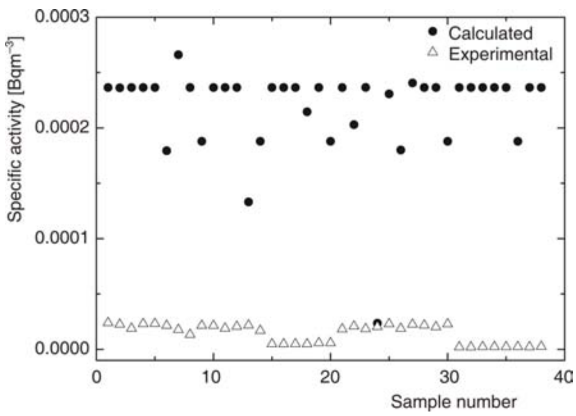

Figure 5. Dependency of the experimentally obtained (circles) and calculated (triangles) ^{137}Cs average weekly specific activities for samples measured during 2010

connected to nodes in the hidden layer (bias 1) and in the output layer (bias 2).

Figure 5 presents values of experimentally obtained and calculated specific activities of ^{137}Cs that were used in the network testing process. It is obvious that predicted values are much lower than actually measured and the correlation coefficient is rather low (r = 0.20).

Figure 6 presents calculated and experimentally obtained values of the ^{137}Cs specific activities as the

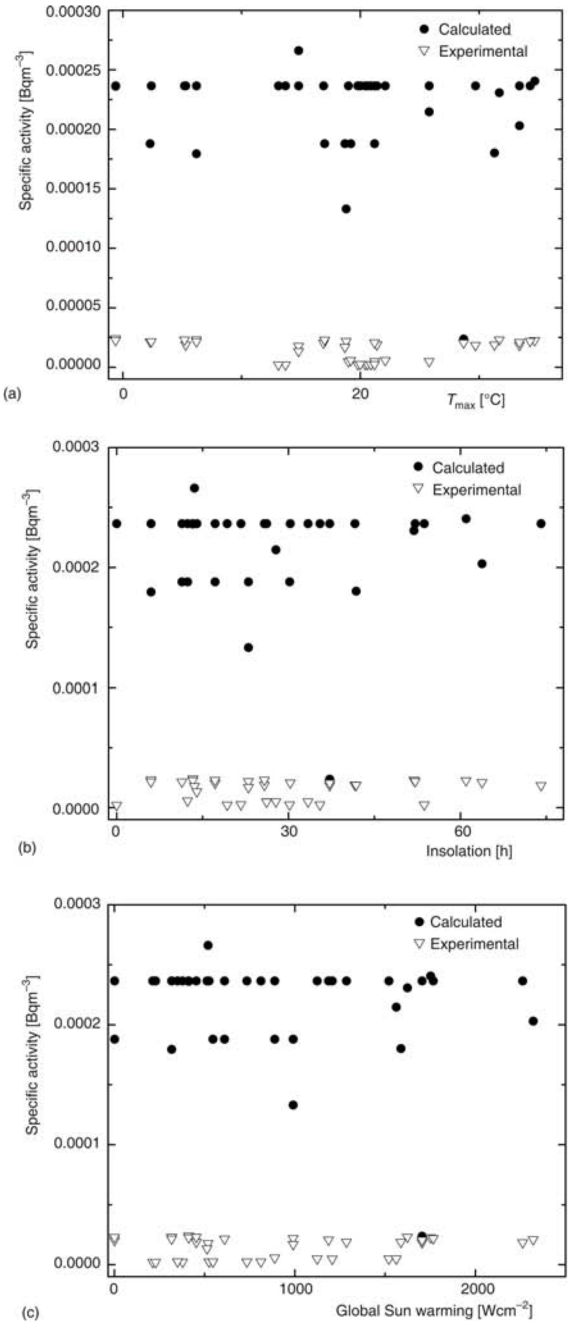

Figure 6. The dependency of experimentally obtained (triangles) and calculated (circles) ^{137}Cs average weekly specific activities as a function of meteorological parameters: (a) maximum air temperature, (b) insolation, and (c) global Sun warming

Table 2. Correlation coefficients of meteorological parameters of air and specific activities experimentally obtained and meteorological parameters of air and calculated specific activities

Meteorological parameters of air	Correlation coefficients of experimentally obtained specific activity and meteorological parameters	Correlation coefficients of calculated specific activity and meteorological parameters
Insolation [h]	0.10	–0.01
Global sun warming [Wcm^{-2}]	0.17	–0.19
Maximum temperature [°C]	–0.15	–0.14

function of individual meteorological parameters, obtained during 2010, while tab. 2 shows the correlation coefficients between these parameters and experimentally obtained and calculated specific activities, for the same period. Values of correlation coefficients are low. Good prediction was obtained only for the maximum temperature while for the other two parameters even the sign of correlation is different between measured and predicted values.

DISCUSSION

The specific activities of ^{137}Cs obtained in this study were in the range 0.09-2.32 10^{-5} Bq/m^3 (see fig. 1). For the 2004-2009 period, Todorović *et al.*, [27] reported that values of ^{137}Cs specific activity were mostly below the detection limit of their technique (10^{-6} Bq/m^3). Another publication [28] reports that during the 1990s these values ranged from 10^{-6}-10^{-5} Bq/m^3, which was in the same range as in different parts of Europe [29, 30]. The maximum values that were observed in late spring/early summer and in winter have been observed by others as well and can be explained by the stratosphere-troposphere exchange (spring/summer) or the soil dust re-suspension in air from the Chernobyl fallout (winter) [31, 32]. Considering the relatively long half-life of ^{137}Cs and relatively infrequent nuclear accidents, its specific activity can remain the same for a long period of time, which was indeed the case until the nuclear accident in Japan in 2011. During the Fukushima accident, reported activity values of ^{137}Cs in Belgrade ranged from 4 to 16 10^{-5} Bq/m^3 [33-35].

The overall correlation coefficient between experimentally obtained specific activities and calculated (predicted) values was low (0.20) indicating that our model cannot correctly forecast ^{137}Cs activity. This is a very different situation than for cosmogenic ^{7}Be where a rather good correlation of r = 0.91 has been obtained using the same methodology [20]. This is probably due to the fact that values of all individual correlation coefficients are rather low (tab. 2), and hence their statistical significance is low.

Considering the behaviour of ^{137}Cs and its transfer through the atmosphere, failure in predicting ^{137}Cs specific activities suggests relative independence of local meteorological conditions, *i. e.*, these do not greatly affect the anthropogenic radionuclide ^{137}Cs specific activity. Actually, some authors have stated that there is no significant correlation between them *et all* [36]. It has been suggested [36] that correlation can be improved if sampling is performed on a daily basis instead of on a weekly basis as done here. Even so, it is difficult to believe that this can increase the correlation to the level of reliable forecasting.

Benefits of using PCA/ANN methods in forecasting cannot be seen when correlation coefficients for individual meteorological parameters and specific activities of ^{137}Cs are low. However, this method has been found to be useful in cases of forecasting concentration levels of other airborne radionuclides such as ^{7}Be and ^{3}H and air pollutants [20, 37, 38] and using PCA/ANN seems to be the only way analyse multiple parameters and unknown analytical relationships.

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AUTHOR CONTRIBUTIONS

Principal component analysis was carried out by A. D. Samolov and M. Ž. Daković, neural networks analysis was carried out by A. D. Samolov and S. D. Dragović, while experiments were carried out by A. D. Samolov. All authors analysed and discussed results and wrote the manuscript. The figures were prepared by A. D. Samolov and M. Ž. Daković.

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ПРИМЕНА НЕУРОНСКИХ МРЕЖА ЗА АНАЛИЗУ ПОНАШАЊА ^{137}Cs У УЗОРЦИМА ВАЗДУХА НА ПОДРУЧЈУ ГРАДА БЕОГРАДА

У раду је приказан комбиновани метод примене анализе основних компоненти и неуронских мрежа за предвиђање понашања ^{137}Cs у функцији метеоролошких параметара. Модел је оптимизован и тестиран коришћењем вредности специфичних активности ^{137}Cs добијених стандардном гамаспектрометријском анализом узорака ваздуха, који су сакупљани у периоду 2009-2011. године на територији Београда, и метеоролошких параметара из истог периода. Добијена је мала вредност корелационог коефицијента (r = 0.20) између експерименталних и неуронском мрежом израчунатих вредности специфичних активности ^{137}Cs. Ово показује да неуронска мрежа за случај предвиђања вредности специфичних активности ^{137}Cs, у функцији температуре, инсолације и глобалног Сунчевог загревања не показује добре резултате, што може да се објасни релативном независношћу вредности специфичних активности ^{137}Cs од метеоролошких параметара, а не неефикасношћу вештачких неуронских мрежа.

Кључне речи: неуронска мрежа, гамаспектрометрија, ваздух, ^{137}Cs